INCLINED SUBSTRATE PULSED LASER DEPOSITION OF YBCO THIN FILMS ON POLYCRYSTALLINE AG SUBSTRATES

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ABSTRACT

Films of $YBa_2Cu_3O_{7-x}$ (YBCO) with c-axis orientation were directly deposited on nontextured silver substrates by inclined substrate pulsed laser ablation. The structure of the YBCO films was characterized by X-ray diffraction 2θ-scans, Ω-scans, and pole-figure analysis. A good alignment of the c-axis of the YBCO films was confirmed by the Ω-scans, in which the full width at half maximum of the YBCO(005) was 3.8°. A sharp interface between the YBCO film and Ag substrate was observed by transmission electron microscopy. The surface morphology of the film, examined by scanning electron microscopy, reflected the recrystallization of the Ag substrate. Raman spectroscopy was used to evaluate the quality of the YBCO films. The superconducting transition temperature (T_c) and the critical current density (J_c) of the films were determined by inductive and transport measurements, respectively. T_c = 91 K with a sharp transition and J_c = $2.7 \times 10^5 A/cm^2$ at 77 K in zero external field were achieved on a film with 0.14-μm thickness.

INTRODUCTION

A promising material for future electric power applications is $YBa_2Cu_3O_{7-x}$ (YBCO)-coated conductors [1-4]. Recently, YBCO thin films grown on silver tapes have attracted much interest because of their high superconducting transition temperature (T_c) and high critical current density (J_c). Silver is an ideal substrate candidate due to its compatibility with YBCO and its inertness to oxidation [5-6]. The electrical properties have been improved in the YBCO films by silver doping up to 20 at.% [7-8]. Unlike other substrate materials such as Ni-based alloys, in which buffer layers are necessary to prevent Ni diffusion into the YBCO film, YBCO can be directly deposited on silver substrate without a buffer layer. To achieve high J_c, textured silver sheets are often chosen as substrates. YBCO films grown on these textured substrates have achieved J_c of $\approx 5\times10^5$ A/cm^2 [9-11]. However, a relatively complex process is usually involved in preparing well-textured silver substrate for YBCO deposition.

We have explored direct deposition of YBCO on nontextured silver substrate for simplifying the fabrication process. In our studies, nontextured polycrystalline silver sheet was used as the substrate, and YBCO films were directly deposited on these substrates by inclined substrate pulsed laser deposition (IS-PLD). Inclined substrate deposition is a promising technique for growing biaxially textured films on nontextured substrates [3-4], and improvement in film quality is expected from this technique. This work shows the feasibility for fabricating high-J_c YBCO films on nontextured polycrystalline silver substrate.

EXPERIMENTAL

Inclined substrate PLD was used to grow YBCO films. A schematic diagram of the deposition system is shown in Figure 1. Substrates with silver paste were mounted on a tiltable heater. Different from the conventional arrangement in which the surfaces of the target and the heater were parallel, the heater can be tilted at an angle with respect to the target surface. The substrate inclination angle, α, substrate normal with respect to the evaporation plume direction, was typically set at 55°, which is believed to be a favorable angle to produce (001) oriented textured film. To compare the dependence of α, 35° and 72° were also tested. Nontextured polycrystalline silver coupons, 5×10 mm in dimensions and 0.1 mm in thickness, were used as substrates after mechanical polishing with

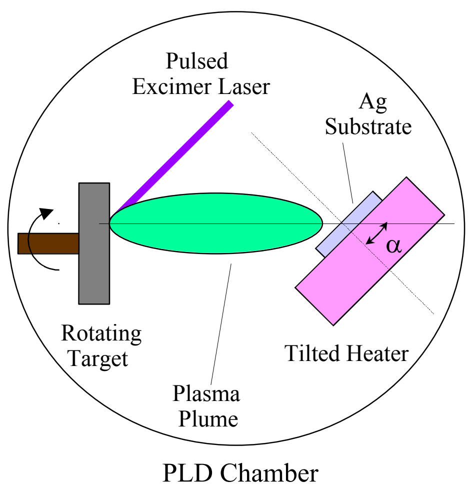


Figure 1. Schematic diagram of inclined substrate pulsed laser deposition system

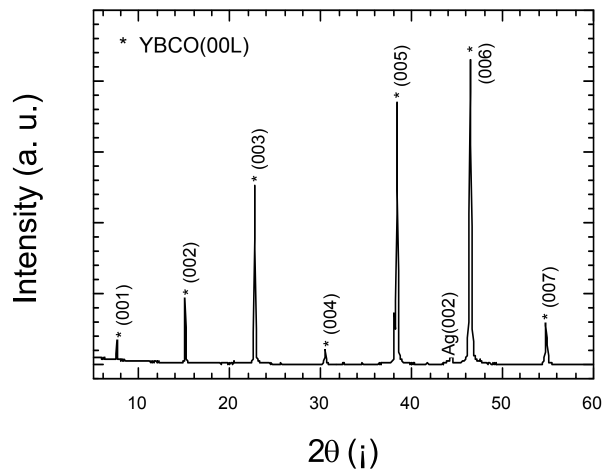
diamond paste up to the 1-μm level. The substrates were heated programmatically to a desired deposition temperature. Substrate temperatures were measured by a thermocouple mounted on the surface of the heater.

An excimer laser (Lambda Physik, Compex 201) with a wavelength of 248 nm and pulse width of 25 ns was used for the deposition. Conditions included a pulse repeat rate of 8 Hz and oxygen pressure of 200 mtorr. The laser was focused at a rotating target with an energy density of 2~3 J/cm^2. A commercial YBCO target (Superconductive Components Inc., purity of 99.999%), 2.5 cm in diameter and 0.64 cm in thickness, was used. The distance between the target and the substrate was 7 cm. The typical thickness of the YBCO films was 0.14 μm.

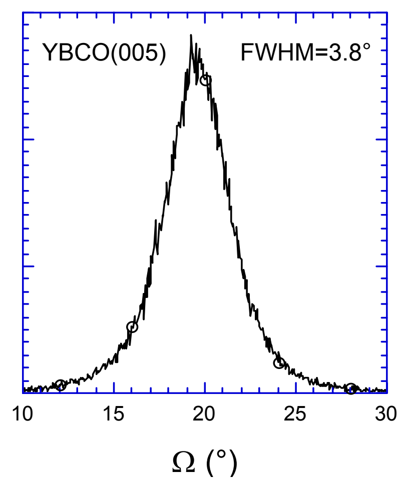
The structures of these films and the substrates, including the out-of-plane and in-plane textures, were examined by X-ray diffraction (XRD) 2θ-scans, Ω-scans, and pole-figure analysis. Transmission electron microscopy (TEM) and scanning electron microscopy (SEM) were used to observe the interface structure and the surface morphology of the samples, respectively. The quality of the films was also characterized by Raman spectral analysis. An inductive method was used to measure T_c. The J_c was determined by the standard four-point method on samples with typical size of 5 × 10 mm using a criterion of 1 μV/cm.

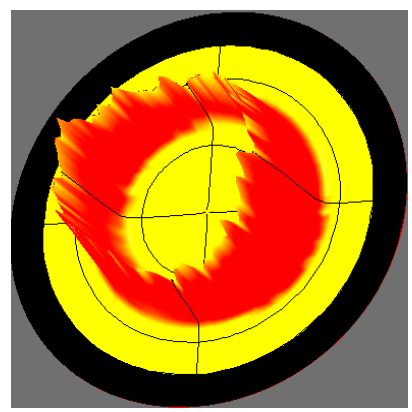
RESULTS AND DISCUSSION

The crystalline structure of the YBCO films grown on polycrystalline silver substrates was determined from the XRD 2θ-scan pattern, shown in Fig. 2(a). This XRD pattern consists of a set of sharp and strong peaks, which have been identified as YBCO(00*l*) peaks. No a-axis orientation peaks were observed in this pattern, indicating good c-axis alignment in the YBCO films. A full- width- at-half- maximum (FWHM) of ≈0.2° was measured for the YBCO(005) peak, suggesting a high-quality crystalline structure. The c-axis alignment was investigated by Ω-scans, shown in Fig. 2(b). The Ω-scans of the YBCO(005) peak gave a FWHM value of 3.8°, indicating a small orientation dispersion of the c-axis. To characterize the in-plane texture, X-ray pole-figure analysis of the YBCO(103) plane was performed, as shown in Fig 2(c). It reveals that the intensity of the diffraction of the selected plane is fairly uniformly distributed over the whole 360° ϕ-angle range, indicating no in-plane texture. This finding is expected from the polycrystalline texture of the substrate.



(a)





(b) **(c)**

Fiure 2. X-ray diffraction patterns of YBCO/Ag films: (a) 2θ-scan, (b) Ω-scan of YBCO(005), and (c) pole-figure of YBCO(103)

Raman spectra were also used to obtain information about the phase integrity, cation disorder, and second-phase formation such as CuO and $BaCuO_2$. Figure 3 shows the Raman spectrum of the as-grown YBCO films, which exhibit a strong Raman band at 340 cm^{-1}, along with a weak band centered at about 500 cm^{-1}. The strong Raman band at ≈ 340 cm^{-1} indicates that a c-axis-oriented YBCO film was formed. The existence of the weak band at around 500 cm^{-1} suggests minimal c-axis misalignment of the YBCO film [12-13], in this case, in-plane texture disorder, to some extent. Lack of other bands suggests that the YBCO film has a stoichiometric composition.

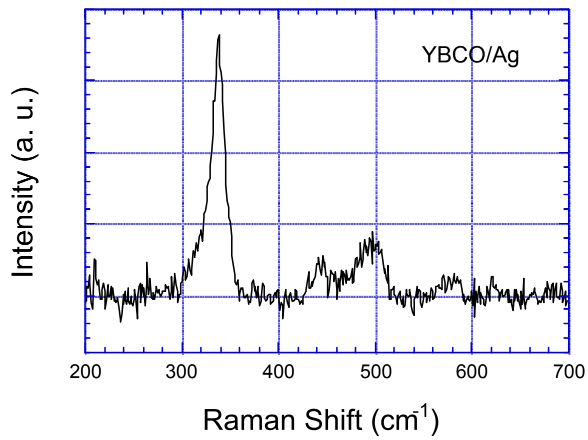


Figure 3. Raman spectrum of YBCO films grown on the Ag substrates

The T_c of the YBCO films grown on silver substrates was measured by an inductive method. Figure 4 shows a typical superconducting transition temperature pattern of the YBCO films. T_c values of 91 K with narrow transition widths of 3.0 K were achieved for the YBCO film deposited on silver substrate at

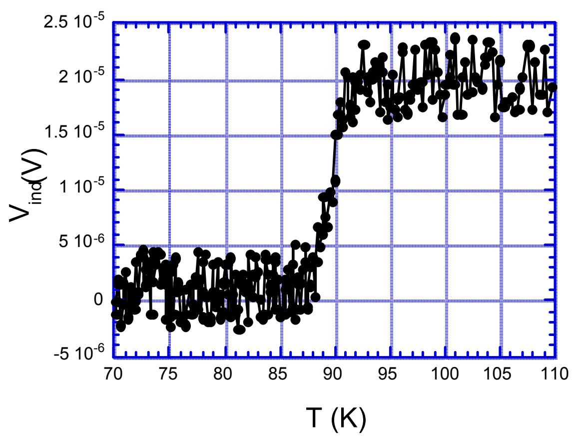


Figure 4. T_c pattern of YBCO/Ag films measured by inductive method.

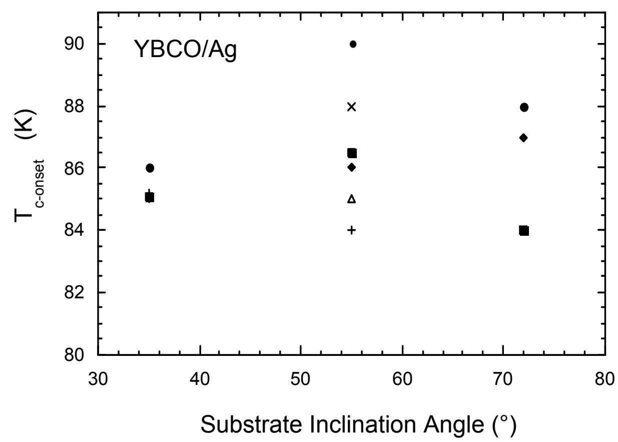


Figure 5. T_c of YBCO films as function of substrate inclination angle

755 °C with a substrate inclination angle of 55°. The onset T_c values and the transition widths of the YBCO films deposited at inclined substrate angles of 35°, 55°, and 72° covered a range, but the best values were obtained on samples deposited with an inclined substrate angle of 55°. Figure 5 shows the T_c distribution over different inclined substrate angles. The J_c of the YBCO films was measured by the four-point transport method over an entire 5×10 mm sample. Before the J_c measurement, the YBCO/Ag samples were coated with a silver layer of about 2-μm thickness by e-beam evaporation and then annealed in flowing ultra-high purity oxygen at 400 °C for 0.5 h. A J_c of ≈2.7 × 10^5 A/cm^2 was achieved at 77 K with zero external field for a 0.14-μm-thick YBCO film deposited at 755 °C with an inclination angle of 55°. The high J_c values of the samples may be partly attributed to the possibly improved microstructure of the films and the enhanced surface mobility of the deposited atoms due to the IS-IS-PLD process. Further studies concerning the growth of YBCO films on Ag substrates are under way.

The structure of the Ag substrates was also examined by XRD. Figure 6(a) shows the 2θ-scan pattern of the silver substrates. More than three peaks were found in the region of 2θ = 25-85° in both the as-polished and the post-deposition substrates, suggesting a polycrystalline structure for the substrates. The FWHMs of the peaks from the pattern of the post-deposition substrate were much smaller than those of the as-polished one, indicating the size of the crystalline grains of the as-polished substrates were much smaller than those of the post-deposition substrates. The orientation distribution of the grains was examined with X-ray diffraction Ω-scans. Figure 6(b) shows the Ω-scan of the Ag(200) plane. A broad peak with FWHM=13° was observed for the as-polished substrates, indicating a poor alignment and continual orientation divergence of the Ag(200) plane. For the post-deposition substrate, several peaks with different intensities and FWHM values emerged in the pattern, suggesting a discontinuous orientation of the Ag(200) plane. Similar patterns of discontinuous orientation were also observed for the other lattice planes of the substrates. The transition of the orientation distribution of Ag grains from continuous to discontinuous may have contributed to the recrystallization during the deposition.

TEM observation revealed a clear, sharp interface for the YBCO/Ag sample, as illustrated in Fig 7(a). Figure 7(b) is a selected area electron diffraction pattern

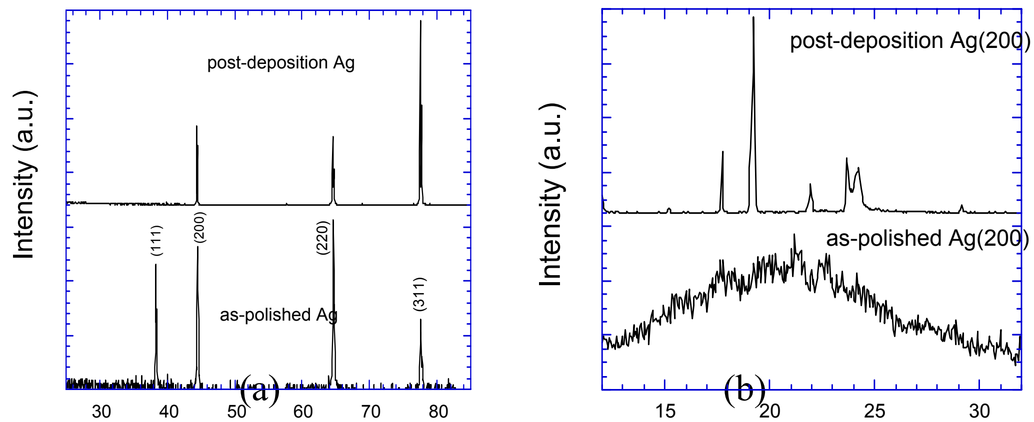
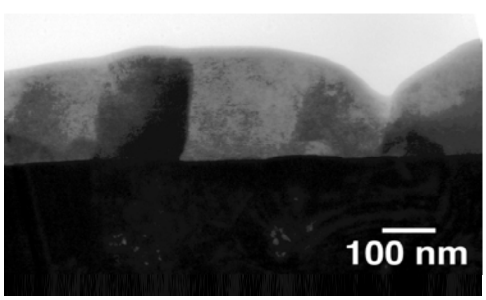


Figure 6. XRD of Ag substrates: (a) 2θ-scans; (b) Ω-scans of Ag(200)

from the interface shown in Fig. 7(a) with the Ag oriented along the [001] zone axis. Further studies on the orientation relationship of the YBCO films over the



(a) (b)

Figure 7. TEM investigation of YBCO/Ag interface: (a) TEM image; (b) electron diffraction pattern

Ag substrate in the interface are under way.

The surface morphology of the YBCO films was examined by SEM. In the low magnification mode, different irregular-mosaic-shaped areas were observed. These areas have clear, sharp borders, but different contrasts and micromorphologies. They can be categorized by contrast to black areas, bright areas, and some intermediate areas, as illustrated in Fig. 8. The black areas have a denser surface and better connection among micrograins, along with a few short-bar-like grains randomly distributed. The bright areas have a less dense surface with some short-bar-like grains lying along a particular direction. However, energy dispersion spectral analysis showed no obvious compositional difference among these areas.

This surface morphology may directly relate to the substrate morphology. On a structurally and compositionally uniform substrate, a uniform film at the microscopic scale is expected to grow, since other growth parameters in PLD are all macroscopic quantities. Since silver has a face-centered cubic crystalline structure and the recrystallization temperature is low, the silver substrate is expected to undergo recrystallization, and large grains may form during the film

Figure 8. SEM image of the as-grown YBCO films in low magnification mode

deposition [5-6]. This may influence the morphology of the deposited YBCO films. To find the exact correlation between the morphology of the films and the orientation of the silver grains, as well as the film formation mechanism, more work is needed.

CONCLUSIONS

YBCO films with c-axis orientation were directly grown on polycrystalline silver substrates by inclined substrate pulsed laser deposition. X-ray pole-figure measurement showed that the in-plane texture of the YBCO films was randomly distributed. Raman spectroscopy confirmed the c-axis orientation and the stoichiometric composition of the YBCO film. Recrystallization of the substrates during deposition was confirmed by XRD Ω-scan. TEM revealed a sharp interface between the YBCO film and the Ag substrate. SEM revealed dense and smooth YBCO films with areas having a variety of irregular-mosaic-shaped morphologies. This effect was attributed to the recrystallization of the substrate. An as-grown YBCO film with thickness of 0.14 μm achieved onset T_c of 91 K, transition width of 3.0 K, and J_c of 2.7×10^5A/cm^2. These studies have demonstrated a promising approach for fabricating high-T_c and high-J_c YBCO films in a simple and economic way for practical application.

ACKNOWLEDGMENT

The authors would like to thank Dr. D. J. Miller for his XRD pole-figure measurement of the YBCO film samples. This work is supported by the U.S. Department of Energy (DOE), Energy Efficiency and Renewable Energy, as part of a DOE program to develop electric power technology, under Contract W-31-109-Eng-38.

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